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Invited paper

Remoteness from sources of persistent organic pollutants in the multi-media global environment☆

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ABSTRACT

Quantifying the remoteness from sources of persistent organic pollutants (POPs) can inform the design of monitoring studies and the interpretation of measurement data. Previous work on quantifying remoteness has not explicitly considered partitioning between the gas phase and aerosols, and between the atmosphere and the Earth's surface. The objective of this study is to present a metric of remoteness for POPs transported through the atmosphere calculated with a global multimedia fate model, BETR-Research. We calculated the remoteness of regions covering the entire globe from emission sources distributed according to light emissions, and taking into account the multimedia partitioning properties of chemicals and using averaged global climate data. Remoteness for hypothetical chemicals with distinct partitioning properties (volatile, semi-volatile, hydrophilic, low-volatility) and having two different half-lives in air (60-day and 2-day) are presented. Differences in remoteness distribution among the hypothetical chemicals are most pronounced in scenarios assuming 60-day half-life in air. In scenarios with a 2-day half-life in air, degradation dominates over wet and dry deposition processes as a pathway for atmospheric removal of all chemicals except the low-volatility chemical. The remoteness distribution of the low-volatility chemical is strongly dependent on assumptions about degradability on atmospheric aerosols. Calculations that considered seasonal variability in temperature, hydroxyl radical concentrations in the atmosphere and global atmospheric and oceanic circulation patterns indicate that variability in hydroxyl radical concentrations largely determines the seasonal variability of remoteness. Concentrations of polybrominated diphenyl ethers (PBDEs) measured in tree bark from around the world are more highly correlated with remoteness calculated using our methods than with proximity to human population, and we see considerable potential to apply remoteness calculations for interpretation of monitoring data collected under programs such as the Stockholm Convention Global Monitoring Plan.

1. Introduction

The potential of a chemical to pose an environmental and/or human health risk as a result of long-range transport is a defining property of a persistent organic pollutant (POP). As such, long-range transport potential is identified in the Stockholm Convention as one of the screening criteria for determining whether a substance should be classified as a POP, and subject to global regulation. According to Annex D of the convention text (S.C, 2009), measured levels of a chemical "in locations distant from its release" should be supplied as evidence of a chemical's long-range transport potential during the screening process. Therefore, in the planning and design of measurement campaigns, and also, when analyzing measurement data from monitoring networks, it is critical to determine how distant or remote sampling locations are from pollution sources.

In a pioneering study, von Waldow et al. (2010) introduced a quantitative metric of the remoteness of regions at a global or regional scale from emissions of pollutants to the atmosphere, called remoteness index. Von Waldow et al.'s (2010) remoteness index is calculated from atmospheric transport modeling of a suite of volatile tracers with different atmospheric half-lives emitted at a constant rate on a specified geographical distribution. The results of the tracer modeling are fit to a non-linear function that provides a

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location-specific parameter that indicates the remoteness of locations in the model domain from the emissions. Westgate et al. (2010) subsequently proposed three methods to quantify the remoteness of air sampling sites. They define pertingency index values calculated for sampling sites as a measure of the proximity (1/remoteness) of the sites to spatially distributed emissions. Two of the methods proposed by Westgate et al. (2010) are based on the geographical distance of sources from sampling sites, and one method uses Lagrangian trajectory modeling. They applied these methods to determine the proximity of the sampling sites in the Global Atmospheric Passive Sampling (GAPS) study to estimated global emissions of polycyclic aromatic hydrocarbons (PAHs). Similar methods using Lagrangian trajectory modeling to quantify the remoteness of sampling sites in studies were later used by Westgate and Wania (2011) and Westgate et al. (2013).

In the approach proposed by Westgate et al. (2010), the location of a sampling site and also the specific time period for sampling are required to obtain the air-mass trajectories that end at the site. While, in principle, the whole globe can be divided as a collection of cells and the air-mass back trajectories for all cells can be calculated, it is more practical to use this method to quantify remoteness of a limited number of predetermined sampling sites to a given emissions distribution to avoid extensive computations. Von Waldow et al.'s (2010) method, on the other hand, used a simplified global atmospheric circulation model to calculate long-term average circulation patterns at steady-state as the basis of the remoteness index. Von Waldow et al.'s (2010) method, therefore, produces time-independent remoteness values unlike the methods based on air-mass back-trajectory calculations.

Neither the von Waldow et al. (2010) nor the Westgate et al. (2010) approaches to quantify remoteness take into account the properties of specific pollutants. In this study, we propose a method to quantify remoteness from global sources of POPs that considers the multimedia fate and transport properties of chemical pollutants. We use a global multimedia fate model to derive remoteness values of regions covering the entire globe for a set of hypothetical pollutants assuming a representative generic global emissions distribution. Running the fate model in steady-state mode yields long-term average remoteness values similar to the von Waldow et al. (2010) approach. However the global multimedia fate model is capable of dynamic simulations, and therefore, seasonal variations in the remoteness of locations can be analyzed with our method when time-dependent remoteness values are of interest.

Our method of quantifying remoteness is based on the transfer efficiency concept introduced by MacLeod and Mackay (2004). Transfer efficiency is a model-based metric of the contribution of emissions distributed in a defined geographical pattern to the flux of chemical that reaches a specific receptor area. In MacLeod and Mackay's (2004) analysis, the source area was the continent of North America and the receptor area was the Great Lakes. In this study, the whole globe is considered as both the source and the receptor.

This study presents an investigation of the relationship between the multimedia partitioning properties of pollutants and the remoteness of regions from pollution sources. Hypothetical POPs that represent different categories of partitioning properties are used in the analysis. A case study with polybrominated diphenyl ethers (PBDEs) is included as an evaluation of the methodology. The proposed approach for quantifying remoteness allowed us to conduct a critical analysis of the 2-day-half-life-in-air criteria that is used to screen for long-range transport potential in the Stockholm Convention and under various national regulations as it is applied to pollutants with different multimedia partitioning properties.

2. Methods

2.1. Global multimedia fate model – BETR-Global

We used the global multimedia fate model BETR-Global (MacLeod et al., 2011) to calculate the global transfer efficiency values for the hypothetical and real organic chemicals analyzed in this study. BETR-Global is a member of the Berkeley-Trent (BETR) family of spatially resolved multimedia fate models. In BETR models, the environment is conceptualized as a collection of interconnected regional mass-balance models. Each region has seven compartments: upper atmosphere, lower atmosphere, vegetation, freshwater, ocean, soil, and freshwater sediments. The regions are connected by bi-directional flows of air and water between the atmosphere, ocean and fresh water compartments. Chemical mass balance equations for each of the compartments in each of the model regions are defined, and numerical solution of the resulting set of equations provides the spatial and temporal distribution of the simulated chemical.

Since its first introduction (MacLeod et al., 2005), BETR-Global has been applied to analyze many different global-scale chemical pollution problems (e.g. Armitage et al., 2009a; Armitage et al. 2009b; Gusev et al., 2010; Lamon et al., 2009; Li et al., 2015; Wöhrnschimmel et al., 2012). These applications have built confidence in the model structure and assumptions by conducting model evaluations where modeled and measured concentrations of chemicals are compared. Lamon et al.'s (2009) study showed satisfactory agreement between the dynamic model results and long-term monitoring data for globally distributed concentrations of PCB 28 and PCB 153 in air. Wöhrnschimmel et al. (2012) modeled the global fate of α- and β-HCH and evaluated the model performance by comparing the model results with measured concentrations in air and ocean water. Recently, two new software implementations of BETR-Global has been published (MacLeod et al., 2011). BETR-Global 2.0 is coded in Visual Basic for Applications and runs as a Microsoft Excel macro (https://sites.google.com/site/betrglobal). BETR-Research is a re-implementation of BETR-Global in the Python programming language (http://betrs.sourceforge.net). BETR-Research is an open-source project aimed at researchers who would like to modify the model code and add new capabilities to the model according to their specific research interests. Some of the modifications to BETR-Research since its first introduction include increasing the resolution of the global data sets to model the global environment on $3.75^\circ \times 3.75^\circ$ spatial resolution (in addition to the base resolution of $15^\circ \times 15^\circ$), implementing a fast numerical solver, and adding new code to track mass fluxes between compartments. In this study, the newest version of BETR-Research that allows fast simulations with high spatial resolution was used. Code introduced to the model to track mass fluxes between compartments (Wöhrnschimmel et al., 2013) was critical in calculating the transfer efficiency values.

2.2. Quantifying remoteness

Our proposed multimedia remoteness measure is based on the global distribution of transfer efficiency values calculated by the BETR-Research model. The definition of transfer efficiency used in this study is a slight modification of the definition given by MacLeod and Mackay (2004). In this study, transfer efficiency is calculated by dividing the rate of contaminant flux to a selected target environmental compartment (including emissions) by the rate of global emissions:

$$\text{Transfer Efficiency (TE)} = \frac{\text{Rate of contaminant flux to the target environmental compartment (mol/h)}}{\text{Rate of global emissions (mol/h)}} \quad (1)$$

Our remoteness measure is then calculated by transforming the TE values so that lower values indicate less remoteness while higher values indicate more remoteness. This is achieved by defining remoteness as equal to −log(TE). The resultant dimensionless remoteness metric is called pTE:

$$\text{pTE} = -\log(\text{TE}) \quad (2)$$

In this study, the focus is on POPs that are emitted into, transported through and measured in the atmosphere. Therefore, the target environmental compartment in Eq. (1) is the lower atmosphere compartment, and the transfer efficiency to a model region is calculated by summing all the atmospheric fluxes into the lower atmosphere compartment of that region, i.e., the sum of the input fluxes from the lower atmosphere compartments of the neighboring regions, input flux from the upper atmosphere of the model region, and the direct emissions to the lower air compartment. This approach is compatible with the remoteness concept that has been proposed in previous studies (von Waldow et al., 2010; Westgate et al., 2010), which were also primarily interested in atmospheric pollutants. However, in principle, the remoteness measure proposed in this study (pTE) can be used to quantify remoteness of any model compartment and could consider transport in water as well as air.

2.3. Simulation experiments

2.3.1. Remoteness analysis with hypothetical chemicals

Wania (2006) and Gouin and Wania (2007) proposed a categorization of chemicals according to their air–water and octanol–air partition coefficients, K_{AW} and K_{OA}, respectively. This categorization defines four types of POPs according to their global transport behavior (Fig. S1). The "flyers" are volatile chemicals that do not deposit onto surface compartments. At the other extreme, "single-hoppers" have low volatility and are predominantly associated with aerosols in the atmosphere and irreversibly deposit onto surface compartments. Between these two extremes are "multiple hoppers" that are semi-volatile chemicals that are readily exchanged between the atmosphere and the surface compartments, and "swimmers" that tend to be present in the water phase and transport via oceans. In this study, four hypothetical chemicals (volatile, semi-volatile, hydrophilic and low-volatility) that belong distinctly to each of these categories (flyers, multiple-hoppers, swimmers and single hoppers, respectively) are defined by assigning appropriate partitioning properties between air, water and octanol. Table 1 lists the partition coefficient values assigned to each of the hypothetical chemicals.

Annex D of the Stockholm Convention (S.C, 2009) states that for an organic pollutant to be classified as persistent, its half-life in water should be longer than 2 months, or its half-life in soil and/or sediment should be longer than 6 months. In this study, all the hypothetical chemicals are assigned these limit values as their half-lives in water, soil and sediment compartments. Half-life in vegetation is assumed to be the same as half-life in water. We considered two different scenarios for degradation half-lives in air, where chemical degradation in air is assumed to occur only by reaction with hydroxyl radicals. In the first scenario, the hypothetical chemicals are assumed to have a 60-day half-life in air when exposed to hydroxyl radicals in the gas phase at global-average concentrations. This scenario simulates the properties of POPs that are very persistent in air (e.g. similar to hexocholorobenzene). In a second scenario, the hypothetical chemicals have a 2-day half-life in air when exposed to hydroxyl radicals in the gas phase at global-average concentrations, which is the limit value given in the Stockholm Convention (S.C, 2009, Annex D) when screening for chemicals with long-range transport potential. The half-lives of hypothetical chemicals used in model simulations to analyze our proposed remoteness metric are given in Table 1.

2.3.2. Remoteness distribution of PBDEs

Salamova and Hites (2013) reported concentrations of PBDEs in tree bark from 12 locations around the globe. They showed a correlation between concentrations of PBDEs in tree bark and human population in nearby areas. As shown in Table 1, PBDEs have properties that place them between the semi-volatile and the low-volatility hypothetical chemicals considered in our calculations. Therefore the dataset from Salamova and Hites (2013) offers an opportunity to evaluate our pTE calculations by investigating the correlation between concentrations of PBDEs in tree bark and remoteness values calculated for the sampling sites. In Salamova and Hites (2013), on average, 80% of total PBDEs measured in tree-bark samples was comprised of BDE-47, -99, and -209. We calculated the global remoteness distributions of these three PBDE congeners using the property values in Table 1 and assuming they are emitted to air according to the night-light emission scenario defined below.

Table 1
Properties of hypothetical chemicals and PBDEs used in model simulations.

	(*Flyer*) volatile	(*Multi-hopper*) semi-volatile	(*Swimmer*) hydrophilic	(*Single-hopper*) low-volatility	BDE-47*	BDE-99*	BDE-209*
			Partition coefficients				
log K_{OW}	6	6	2	8	6.53	7	9.97
log K_{AW}	3	−2	−4	−4	−3.12	−3.37	−4.81
log K_{OA}	3	8	6	12	10.4	11.3	16.8
			Degradation half-lives (days)				
in Air			60 (1st scenario); 2 (2nd scenario)		11	19	318
in Water			60		192	354	1583
in Vegetation			60		192	354	1583
in Soil			180		385	708	3167
in Sediment			180		1155	2125	9500

*: BDE-47, -99, -209 properties are compiled from Schenker et al. (2008).

2.3.3. Emission scenario

For all calculations presented here we used an emission scenario that assumes the global distribution of pollutant emissions follows the same pattern as nighttime light emissions into space. This approach was originally proposed by von Waldow et al. (2010) and was also used in subsequent studies that applied BETR-Global to model pollutants whose usage is related with economic activity and population density (e.g. MacLeod et al., 2011; Wöhrnschimmel et al., 2013). Fig. S2 in the Supporting Information shows the global emissions distribution used in the simulations.

3. Results

3.1. Global remoteness distributions of POPs

Fig. 1 shows the global distribution of pTE for the four hypothetical chemicals when the half-life in air is 60 days. The minimum, the maximum and the mean pTE values for each type of chemical are specified in the figure, and histograms illustrate the frequency of occurrence of different pTE values for each hypothetical chemical. The distribution of remoteness follows the distribution of emissions and also the global atmospheric circulation patterns. Visual comparison of pTE distributions and the histograms indicate that a higher fraction of the global atmosphere at surface level is more remote for the hydrophilic and low-volatility chemicals compared to the volatile and semi-volatile chemicals. The minimum, the maximum and the mean pTE values increase in the order of Volatile < Semi-Volatile < Hydrophilic < Low-Volatility. The same trend in the distribution of pTE values can be seen in the histograms.

3.2. Correlation of remoteness values and tree-bark concentrations for PBDEs

The global remoteness distribution maps for PBDE-47, PBDE-99 and PBDE-209, and the average remoteness distribution map for PBDEs are plotted in Figs. S3 and S4, respectively, and the pTE values at the sampling locations of Salamova and Hites (2013) are given in Table S1. Atmospheric transport of the PBDEs and the low volatility chemical shown in Fig. 1 is mostly determined by the model's description of aerosol transport, so the remoteness distributions of these chemicals are very similar. The correlations between concentrations of PBDEs in tree bark with population and with the pTE values of the sampling locations are shown in Fig. 2. The correlation of the logarithm of concentration values with pTE (r^2 = 0.48, p = 0.012) is stronger than it is with population (r^2 = 0.35, p = 0.045).

3.3. Remoteness for persistent organic pollutants with 2-day half-life in air

Similarly to the scenario for the 60-day half-life in air, the distribution of remoteness in the 2-day half-life in air scenario for the hypothetical chemicals follows the distribution of emissions and also the global atmospheric circulation patterns (Fig. 3). However, the maximum pTE values for volatile, semi-volatile and hydrophilic pollutants are much higher for this scenario, i.e., 8.6, 8.6, and 8.8, respectively (note the different pTE scale in Fig. 3 compared to Fig. 1). The histograms and maps of pTE distributions in the 2-day half-life scenario demonstrate that the pTE value distribution is nearly identical for the volatile and the semi-volatile chemicals,

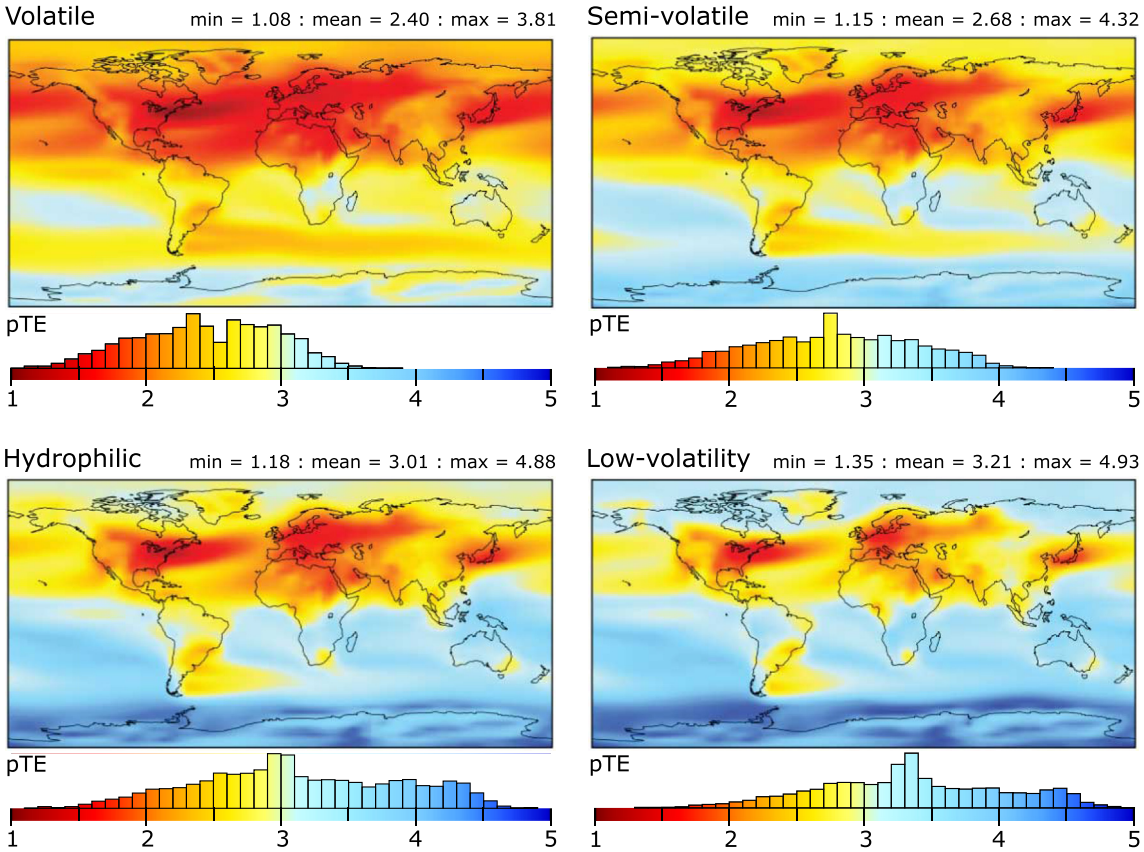

Fig. 1. Global distribution of remoteness (pTE) obtained from steady-state simulations of four hypothetical chemicals when half life in air is 60 days. Histograms illustrate the frequency of occurrence of specified pTE values for each of the four chemicals.

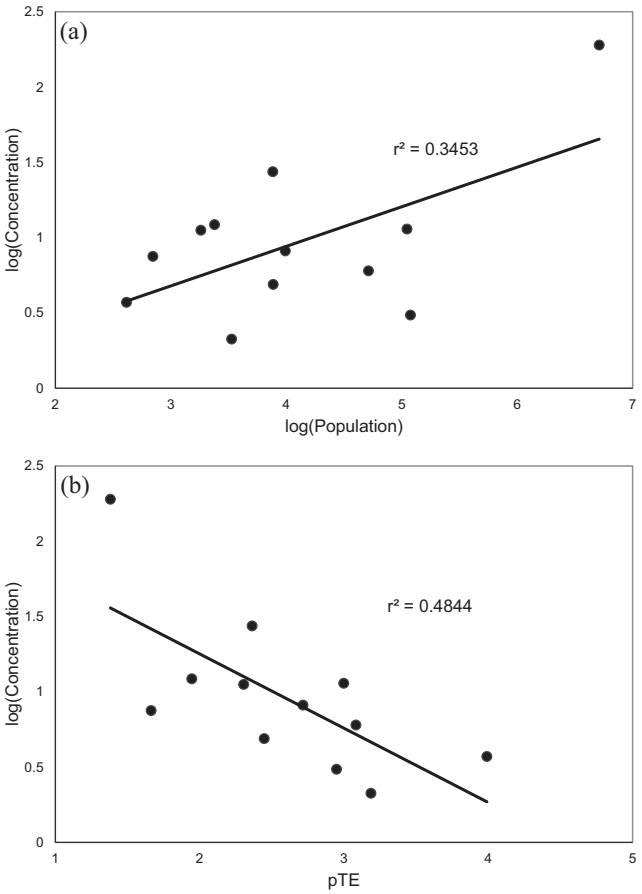

Fig. 2. Regressions of the concentrations of total PBDE in tree-bark samples measured by Salamova and Hites (2013) with (a) population ($r^2 = 0.35$, $p = 0.045$); and (b) pTE ($r^2 = 0.48$, $p = 0.012$).

while the distribution of pTE values for the hydrophilic chemical includes higher values.

In contrast to the other three chemicals, the maximum pTE value simulated for the low-volatility chemical in the 2-day half-life in air scenario is 5 (Fig. 3), which is nearly identical to the maximum pTE value of 4.9 in the 60-day half-life in air scenario illustrated in Fig. 1. Thus, in the 2-day half-life scenario locations far from source areas in the global environment are less remote for the low-volatility chemical than they are for the other 3 substances. Comparing Figs. 1 and 3 and taking note of the different scales indicates that when the 60-day half-life in air was shortened to 2-days, most locations on the earth became more remote for the volatile, semi-volatile and hydrophilic pollutants. However, the shorter half-life affected the remoteness distribution for the low-volatility chemical much less strongly.

3.4. Effect of atmospheric particle associated degradation on the remoteness of the low-volatility chemical

In BETR-Global, one of the base assumptions is to model atmospheric degradation only in the gas phase. Under this assumption, substances that are sorbed to atmospheric particles are not degraded. The model simulations presented above are based on this assumption, which would most strongly affect the low-volatility chemical that is highly associated with aerosols in the atmosphere. There are high uncertainties associated with calculating the amount of chemical sorbed to aerosols and the degradation of sorbed chemicals (Scheringer, 2009). In order to analyze the sensitivity of the remoteness calculations to the quantification of the degradation rate within the aerosols, we designed a simulation scenario that represents the assumption on the other extreme: degradation rate in aerosols being equal to gas-phase degradation rate. We therefore repeated the model simulations for the 2-day half-life in air scenario, but applied that half-life to chemical present in the bulk air compartment consisting of both the gas phase and aerosols. The resultant pTE distribution for the low-volatility chemical is compared with the base case in Fig. 4. Remoteness values calculated for the low-volatility chemical when degradation in air is simulated both in the gas phase and the particle phase are more highly variable geographically, and conform more closely to results for the other three hypothetical chemicals. Repeating the simulations assuming degradation in both the gas phase and aerosols does not significantly affect the remoteness distribution for the other hypothetical chemicals that is illustrated in Fig. 3 (Figs. S5 and S6).

3.5. Comparison of the global pTE distribution of the volatile chemical with the remoteness index distribution of von Waldow et al. (2010)

In order to further evaluate the proposed method for quantifying remoteness from pollution sources, the remoteness index distribution obtained by von Waldow et al. (2010) was compared with the pTE distribution for the hypothetical volatile chemical. Fig. 5a is a reproduction of von Waldow et al.'s (2010) remoteness index distribution for the ECON emission scenario, which also used night-light emissions to represent the geographical distribution of emissions of chemicals. von Waldow et al. (2010) presented the remoteness index values after classifying them as deciles of earth's total surface area. In order to facilitate comparison of pTE distributions with remoteness index distributions, we calculated the percentile ranks of model regions according to their pTE values. Fig. 5b–c shows the percentile rank distributions of pTE values of the volatile chemical for the 60-day air half-life scenario and for the 2-day air half-life scenario, respectively. The pTE distribution for the 60-day air half-life case follows a very similar pattern to the remoteness index distribution of von Waldow et al. (2010). In the case of 2-day air half-life scenario, although a similar pattern is still distinguishable, pTE values are less homogenous.

We calculated the correlation between RI and pTE distributions based on their raw values presented in Fig. S7. The Pearson correlation coefficient (r) between RI and pTE values are 0.88 and 0.75 for the 60-day air half-life and the 2-day air half-life cases, respectively, providing a quantitative measure of the similarity between the RI and pTE distributions (Fig. S8).

3.6. Dynamic global remoteness distribution for the volatile chemical

BETR-Global is capable of dynamic simulations, and therefore, seasonally variable remoteness distributions can be obtained by the proposed method. Using the steady-state model simulation results as initial condition, we performed dynamic global simulations for the hypothetical volatile chemical assuming a 2-day half-life in air. The simulation time period was 5 years and the fifth year's results are presented here interpreted in terms of pTE. Fig. 6 shows the seasonal (three-month) averages of the pTE values for the volatile chemical. The summer and the winter months are visually distinguishable as the Arctic and the whole northern hemisphere are less remote during the winter months, which corresponds to the lowest concentrations of hydroxyl radical in the atmosphere of the northern hemisphere.

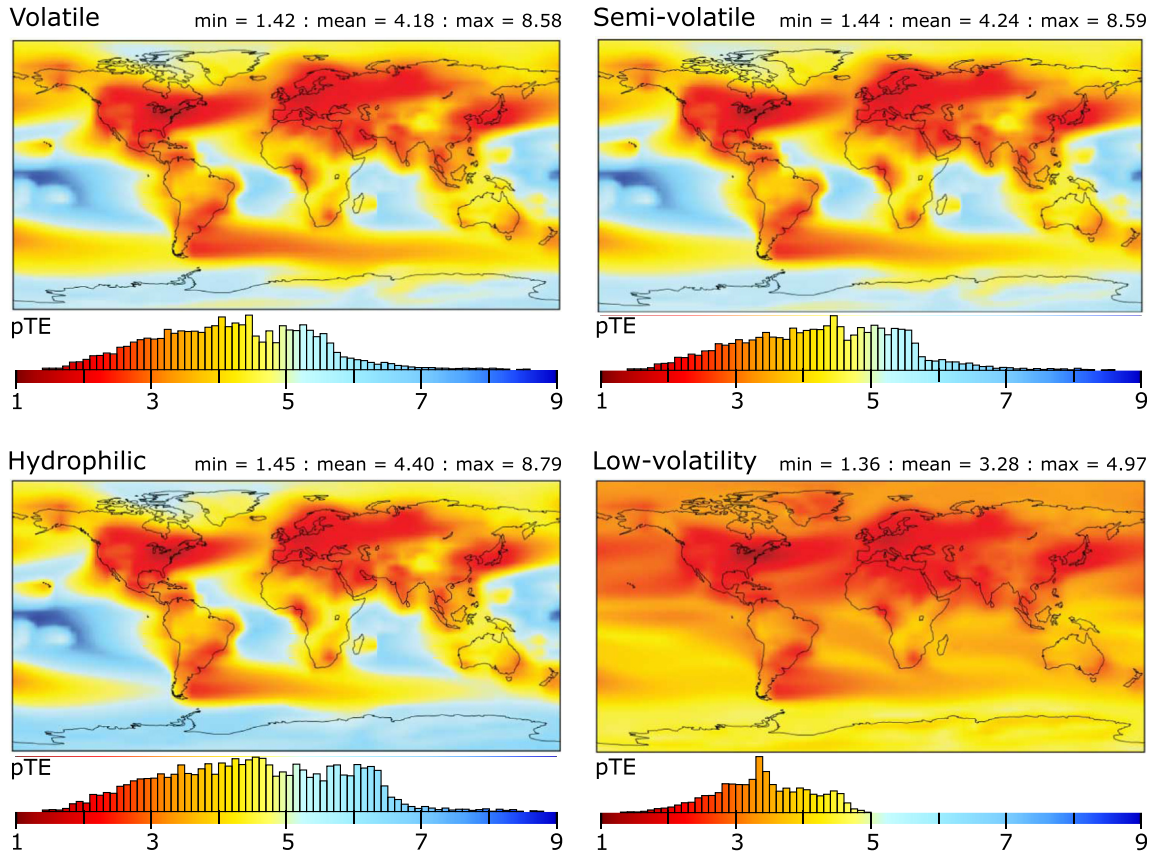

Fig. 3. Global distribution of remoteness (pTE) obtained from steady-state simulations of the four hypothetical chemicals when half life in air is 2 days. Histograms illustrate frequency of occurrence of specified pTE values for each of the four chemicals.

4. Discussion

Remoteness of the global environment calculated for the low-volatility chemical is highly dependent on model assumptions about degradation of chemicals sorbed to aerosols in the atmosphere. It is clear that understanding the atmospheric long-range transport potential of low-volatility pollutants requires an accurate description and parameterization of aerosol-related processes in the model. Scheringer (2009), in a review paper on long-range atmospheric transport, concludes that the degradation rates of particle-associated organic chemicals are “largely unknown” and calls for experimental investigations of the subject. Scheringer (2009) also points out high uncertainties associated with calculating the amount of chemical sorbed to aerosols. BETR-Global calculates aerosol-air partition coefficients from K_{OA} values using the relationship recommended by MacLeod et al. (2010).

Another critical parameter that influences the modeled pTE distributions of the low-volatility chemical is the life-time of aerosols in the atmosphere before being deposited on the surface. We conducted model experiments to determine the average global time scales for removal from atmosphere via wet and dry deposition using hydrophilic and hydrophobic tracers. In BETR-Global, the global average time-scale for the sum of wet and dry deposition is about 3 days for highly hydrophilic pollutants, whereas it is about 19 days for low-volatility chemicals sorbed to aerosols. In our scenarios that assume degradation half-lives in air of 60 days, these deposition processes can compete with degradation in air, and the results reflect the different partitioning behaviors of the hypothetical chemicals. However, when the degradation half-life in air is set to be 2 days, degradation dominates over wet and dry deposition processes as a pathway for atmospheric removal of all chemicals except the low-volatility chemical in the scenario where degradation in air occurs only in the gas phase. In that scenario, deposition is the dominant removal process for the low-volatility chemical. The three PBDE congeners simulated in this study (BDE-47, BDE-99, BDE-209) have different physico-chemical properties (Table 1). However, their partitioning properties are similar to the low-volatility chemical's and they all belong to the single-hopper region in Wania's (2006) chemical space (Fig. S1). The pTE distributions of these PBDE congeners are mostly determined by the lifetime of aerosols in the atmosphere resulting in similar pTE distributions (Fig. S3). Continued research and development is required to improve the description of atmospheric deposition, atmospheric degradation and aerosol-air partitioning processes in BETR-Global, and to provide a better interpretation of the pTE distributions for low-volatility chemicals.

All the analysis presented in this study is focused on pollutants emitted to the atmosphere and the target environmental compartment was chosen as the lower atmosphere. This study design places attention on transport of chemicals from source regions to the atmosphere in remote locations that is in contact with the surface, and it allowed us to evaluate the proposed method within the same context of the previous studies, such as von Waldow et al.'s (2010) remoteness index. However, the pTE method can be used to analyze remoteness of other environmental media (such as water, soil and vegetation) from pollutants emitted to air, water or soil. This type of an analysis is a potential future application of the pTE method introduced in this study.

In the scenario assuming a 60-day half-life in air for the hypothetical chemicals, the volatile chemical is distributed most

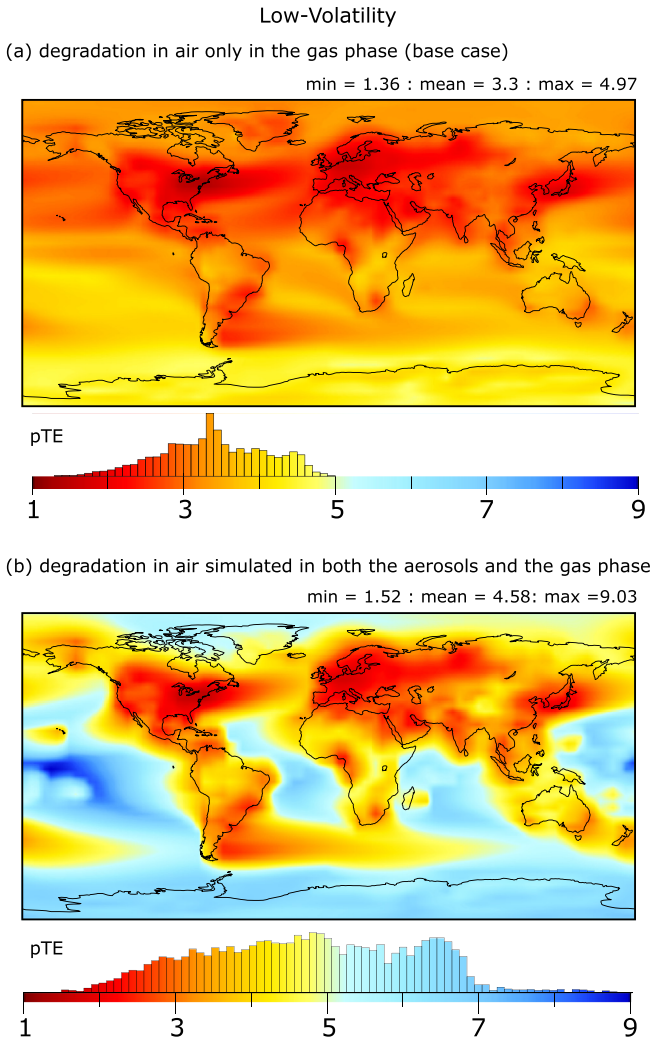

Fig. 4. Global distribution of steady-state pTE values for the low-volatility chemical when half-life in air is 2 days. In case (a), degradation in air is simulated only in the gas phase. In case (b), degradation in air is simulated in both aerosols and the gas phase. Histograms illustrate frequency of occurrence of specified pTE values for each case.

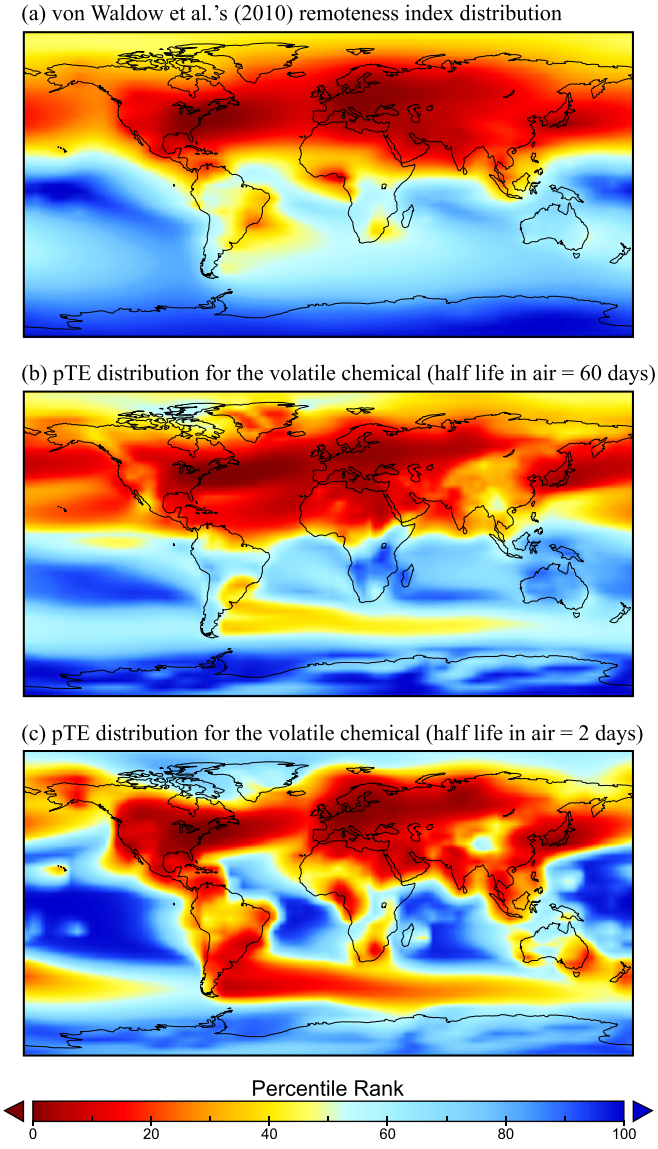

Fig. 5. (a) Percentile ranks of von Waldow et al.'s (2010) remoteness index distribution; and the percentile ranks of pTE distributions for the volatile chemical for both scenarios with (b) 60-day half-life in air, and (c) 2-day half-life in air.

efficiently in the global atmosphere (Fig. 1). In this scenario, even the most remote region for the volatile chemical (pTE = 3.81) is less remote than 23% of the model regions for the low-volatility chemical. The behaviors of the semi-volatile and the hydrophilic chemicals are in-between these two extremes, however, the difference in the remoteness distributions of the volatile and the semi-volatile chemicals is small. The global mean pTE values of the volatile and the semi-volatile chemicals are 2.4 and 2.7 respectively, and only 1.4% of model regions are more remote for the semi-volatile chemical than the most remote region for the volatile chemical. This result suggests that the semi-volatile chemical can be transported to the lower air of regions far from sources nearly as efficiently as the volatile substance. Being more likely to be deposited onto surface compartments of regions after reaching the atmosphere of a remote region, the semi-volatile chemical can cause additional exposure and exposure-related effects. On the other hand, in this model scenario, the remoteness distribution of the hydrophilic chemical is similar to the low-volatility chemical. The global mean pTE values of the hydrophilic and the low-volatility chemicals are 3.0 and 3.2 respectively. And, few model regions are more remote for the low-volatility chemical than the most remote regions for the hydrophilic chemical. When emitted to the atmosphere, the hydrophilic chemical tends to be scavenged by precipitation. Then, its global transport will be mainly through freshwaters and oceans. This behavior is not distinguishable for the hydrophilic chemical in the analysis presented here since this study is focused on transport in the atmosphere and the atmosphere as a target compartment.

The remoteness index (RI) introduced by von Waldow et al. (2010) is an established quantitative measure of remoteness from sources of atmospheric pollutants. RI takes into account the emissions distribution and the global circulation patterns; but it is not dependent on physicochemical properties of pollutants. We compared the pTE distribution of the volatile chemical with the RI distribution for the same emission scenario used in this study (Fig. 5). Although the pTE distribution of the volatile chemical changes when the modeled half-life in air is changed, the resultant pTE distribution maps are comparable indicating that pTE can be used as a quantitative measure of remoteness. Comparison of Fig. 5b–c indicates that when the half-life in air is longer, the pollutant is more homogeneously distributed within the northern

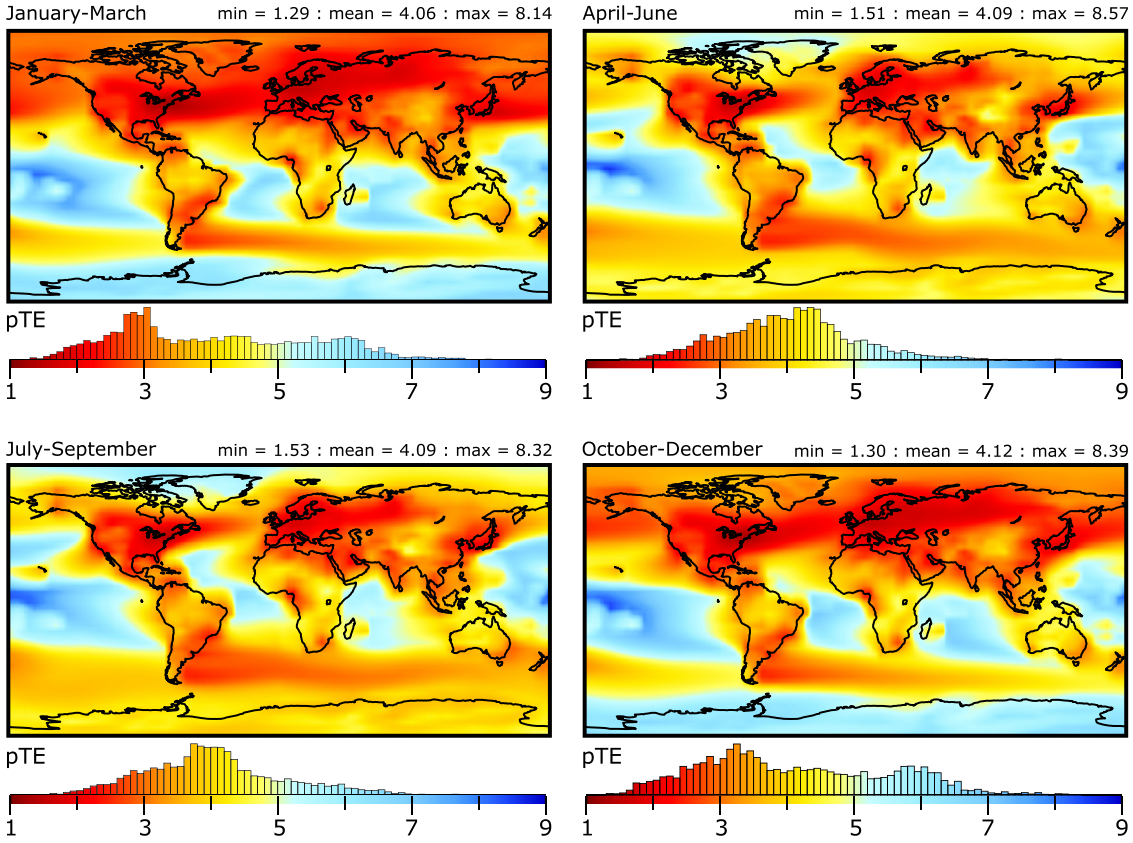

Fig. 6. The dynamic global distribution of pTE values for the volatile chemical assuming an average half life in air of 2-days. Histograms illustrate frequency of occurrence of specified pTE values for each season.

hemisphere since the inter-hemispheric transfers become more significant as a limitation on the global atmospheric transfer efficiencies. Most of the emission sources are located in the northern hemisphere, and the volatile chemical that is not degraded or transferred to the other hemisphere is relatively well-mixed on the hemispheric scales in Fig. 5b (60-day half-life in air). In contrast, in Fig. 5c (2-day half-life in air), there are stronger concentration gradients on hemispheric scales, and the global remoteness distribution closely reflects atmospheric circulation patterns emanating from source regions.

The stronger correlations between PBDE concentrations in tree-bark and pTE compared to population indicate that metrics of remoteness have considerable untapped potential to help interpret measurements of persistent organic contaminants in the global environment. The correlation coefficient ($r^2 = 0.48$) indicates that 48% of the variability in the logarithm of PBDE concentrations in tree bark can be explained by variability in pTE under the assumption that PBDEs are emitted with the same geographical distribution as nighttime light emissions to space, and illustrates that the model's description of transport pathways for these chemicals is reasonable. Using measured concentrations of chemicals in different media to determine correlations with the pTE values calculated for various target environmental compartments other than the atmosphere would be a very interesting analysis. Comparing seasonally varied concentrations with dynamic pTE distributions is also possible and might provide insight into seasonally varying concentrations at remote locations. The remoteness metric has considerable potential to be used in interpretation of monitoring data collected under programs such as the Stockholm Convention Monitoring Plan. In cases such as the one for the PBDEs, where remoteness and measured concentrations are highly correlated, more detailed modeling using alternative emission reduction scenarios will make it possible to formulate strategies to target sources of chemicals for emission reductions and to estimate the effects of these actions on pollutant concentrations at remote locations.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.envpol.2015.12.058.

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